FHI-aims becomes embedded: QM/Me and water splitting

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The development of sustainable and efficient energy conversion processes at interfaces is at the center of the rapidly growing field of basic energy science. This concerns not only desired conversions, such as from solar to chemical energy, but also unavoidable by-products, such as the dissipation of chemical energy into heat. An atomistic understanding of the elementary processes involved is in all cases only just emerging but is likely to question established views and macroscale concepts. In this context, I will review our recent endeavours to advance FHI-aims functionality towards QM/MM embedding. On the one hand, this revolves around a novel approach coined QM/Me that extends the power of embedding techniques to metallic systems. A huge atomistically described bath can thus be included in ab initio molecular dynamics (AIMD) simulations of chemical reactions at model catalyst surfaces. This allows to quantitatively account for the heat dissipation into the phononic system without being riddled by spurious phonon reflections as in conventional supercell AIMD simulations, while simultaneously maintaining the correct description of the metallic band structure of the extended surface. On the other hand this concerns the implementation of pseudopotential functionality to prevent charge leakage in QM/MM models for oxide semiconductor surfaces. In the talk I will sketch the versatility and numerical efficiency of this recently implemented QM/MM-approach in FHI-aims, discussing applications to charge-driven surface redox processes and in particular to direct photocatalytic water splitting.